

Inflection point in the magnetic field dependence of the ordered moment of URu_2Si_2 observed by neutron scattering in fields up to 17 T

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(Dated: 12 August 2002; printed February 6, 2008)

Abstract

We have measured the magnetic field dependence of the ordered antiferromagnetic moment and the magnetic excitations in the heavy-fermion superconductor URu_2Si_2 for fields up to 17 Tesla applied along the tetragonal c axis, using neutron scattering. The decrease of the magnetic intensity of the tiny moment with increasing field does not follow a simple power law, but shows a clear inflection point, indicating that the moment disappears first at the metamagnetic transition at ~ 40 T. This suggests that the moment m is connected to a hidden order parameter ψ which belongs to the same irreducible representation breaking time-reversal symmetry. The magnetic excitation gap at the antiferromagnetic zone center $\mathbf{Q}=(1,0,0)$ increases continuously with increasing field, while that at $\mathbf{Q}=(1.4,0,0)$ is nearly constant. This field dependence is opposite to that of the gap extracted from specific-heat data.

PACS numbers: 75.25.+z, 75.30.Kz, 75.30.Cr, 75.50.Ee, 74.70.Tx

One of the most enigmatic problems in heavy-fermion physics is to reconcile the smallness of the ordered antiferromagnetic (AFM) moment of URu_2Si_2 with the huge λ -anomaly in the specific heat, C_p , at $T_N = 17.5$ K. Neutron [1, 2, 3, 4, 5, 6, 7] and resonant X-ray [8] scattering experiments have shown that a weak static antiferromagnetic dipolar order with the moment along the c axis of the body-centered tetragonal structure (space group I4/mmm) of the heavy-fermion superconductor ($T_c = 1.2$ K) URu_2Si_2 is established below T_N . The magnitude of the moment ($0.03 \mu_B/\text{U-atom}$) is too small to reconcile with the jump in C_p . The specific-heat anomaly suggests the opening of an energy gap over part of the Fermi surface [9, 10]. Inelastic neutron-scattering measurements below T_N show well-defined dispersive magnetic excitations polarized along the c axis, which are consistent with a singlet-singlet crystal-field model [1]. However, such a model predicts an ordered moment much stronger than that observed. Recent reports [11, 12] on that URu_2Si_2 samples are inhomogeneous with a larger moment in small regions are not yet experimentally confirmed and are still a matter of controversy and debate [13]. Also, they do not resolve the discrepancy between the small moment and the specific-heat anomaly.

In this Letter we present the magnetic field dependence of the AFM component of the ordered moment and of the gaps in the magnetic excitation spectrum for fields H up to 17 T applied along the c axis. The high magnetic fields now available for neutron scattering experiments at the HMI made it possible to observe that the magnetic moment in URu_2Si_2 does not vanish at ~ 15 Tesla as was suggested from earlier measurements using a simple power-law extrapolation [4, 14]. Instead, we observe a clear inflection point of the magnetic intensity near a field of 7 T and a finite moment at 17 T. Such a behavior can be obtained if there is a linear coupling between a hidden primary order parameter and the magnetic moment [15]. Inflection points have also been suggested in other theoretical schemes, which will be discussed. The gap Δ in the magnetic excitation spectrum at the antiferromagnetic zone center $\mathbf{Q}=(1,0,0)$ increases strongly with H . The second gap ∇ at $\mathbf{Q}=(1.4,0,0)$ is nearly field independent.

Magnetic fields up to 17 T at low temperatures were obtained by mounting the sample between two dysprosium rods in a 14.5 T superconducting magnet [16]. A high-quality cylindrical URu_2Si_2 single crystal of diameter 6 mm and height 3.8 mm was cut by electroerosion from a larger well-characterized and annealed crystal already used in previous work [14] and mounted with the c axis parallel to the vertical field of the magnet. The neutron scat-

tering experiment was made on the FLEX triple-axis spectrometer of HMI, using pyrolytic graphite as vertically focusing monochromator and horizontally focusing analyzer. Measurement of the magnetic moment was done using the spectrometer in W (RLR) configuration with a wave vector of 1.48 \AA^{-1} . To remove the higher-order harmonic contribution from the monochromator, we used a 10-cm thick liquid-nitrogen cooled Be filter in the incident beam and a graphite filter in the scattered beam, the latter oriented to reflect out the 2nd order harmonics. The inelastic measurements were done with the spectrometer in a “long-chair” (RRL) configuration, a fixed final wave vector of 1.55 \AA^{-1} , and the Be filter in the scattered beam. The energy resolution at elastic energy transfer was 0.14 meV at full width at half maximum. Most of the elastic and inelastic data were collected at a temperature of $T = 2$ K for magnetic fields H between 0 and 17 T.

The integrated intensity of the AFM Bragg peak, which is proportional to the square of the ordered magnetic moment, m^2 , was measured by rocking scans at $\mathbf{Q}=(1,0,0)$, as illustrated in Fig. 1. Measurements above T_N (at high fields) show clearly the absence of any second-order contamination. The finite correlation length is field independent, as reported in earlier measurements [14]. Figure 2 shows the integrated Bragg peak intensity as a function of applied field $H \parallel c$. The figure combines the present data from HMI with earlier measurements at fields up to 12 T from the IN14 spectrometer at the ILL [14], using the field points at 0 and 8 T to scale the results. The magnetic moment decreases with increasing field, but clearly deviates at higher fields from the standard expression

$$m^2 = m_0^2 [1 - (H/H_m)^\gamma] \quad (1)$$

with $\gamma = 2$. Equation (1) describes the magnetic field dependence at $T = 0$ of a (single) order parameter m (the magnetic moment) for a second-order phase transition, and is easily derived in Ginzburg-Landau theory using a free energy of the form

$$F = \alpha(h^2 - t)m^2 + \beta m^4 \quad (2)$$

with $t = 1 - T/T_N$ and $h = H/H_m$, where T_N and H_m are the critical temperature and field for the magnetic order, respectively, and α and β are constants. The critical field H_m was extrapolated to ~ 15 T in earlier low-field work [4, 14]. Modifying the γ exponent (a value of 3/2 was used in Ref. [4]) improves the fit but does not remove the disagreement with the data, which shows a clear inflection point at ~ 7 T.

An inflection point in $m(H)$ was obtained in a model proposed by Shah *et al.* [15], where the magnetic moment m is a secondary order parameter, which is coupled linearly to a hidden primary order parameter ψ , which breaks time reversal symmetry. The magnetic intensity can in this case be written as

$$m^2 = m_0^2 \frac{t - (H/H_C)^2}{[1 + \delta(H/H_C)^2]^2}, \quad (3)$$

where H_C is the metamagnetic field above which the heavy-fermion state is suppressed ($H_C=35.8\text{--}39.4$ T [17]; we use $H_C=40$ T) and δ is a parameter. Shah *et al.* introduces a characteristic field H_M for the magnetic moment through $H_M = H_C/(1 + 2\delta)^{1/2}$. Equation (3) gives an excellent description of the observed field dependence, as illustrated in Fig. 2, with $H_M = 10.4 \pm 0.2$ T as the only adjustable parameter. The linear coupling term $\propto m\psi$ was originally suggested in Ref. 18. Symmetry arguments [15, 18] show that ψ must have the same symmetry as m , i.e. ψ belongs to the A_{2g} irreducible representation (IR). The physical interpretation of ψ is either in terms of triple-spin correlators (octupolar moments or couplings between dipolar and/or quadrupolar moments) or a dipolar moment of the conduction electrons, which have such a rapidly decreasing form factor that the magnetic intensity is negligibly small already at the first observable magnetic Bragg peak [18].

The present finding of an inflection point in $m(H)$ rules out the possibility that the primary order parameter is even under time reversal. Taking the simplest coupling term $m^2\psi^2$ with the right AFM symmetry [15, 18], the field dependence of m is the same as in Eq. (1) [15], and hence in contradiction with the present results. Recent ^{21}Si NMR data suggest in fact that ψ breaks time-reversal symmetry [13]. The existence of an inflection point in $m(H)$ also excludes the scenario with two decoupled order parameters and accidentally close transition temperatures [19], since $m(H)$ would then follow Eq. (1).

While a hidden order parameter that breaks time-reversal symmetry is the most likely scenario, an inflection point in $m(H)$ can also be obtained in a strong coupling model with the magnetic moment as the only order parameter [20]. If the total moment changes between the normal and the magnetically ordered state due to an increase of the magnetic coupling below T_N , then the β term will depend on the external field h and is replaced by $\beta(1 + \eta h^2)$ in Eq. (2), which leads to

$$m^2 = m_0^2 \frac{1 - h^2}{1 + \eta h^2}. \quad (4)$$

Equation (4) provides a good description of the present neutron data, with $\eta = 17.4 \pm 0.7$,

as seen in Fig. 2. However, the model does not resolve the discrepancy between the small moment and the specific-heat anomaly.

Okuno and Miyake [21] have proposed a model which combines the localized character of a the singlet-singlet crystal-field scheme with an itinerant character of the $5f$ electrons. They assume further that there is a perfect nesting at the AFM zone center $\mathbf{Q}=(1,0,0)$. For a suitable parameter set, they obtain an inflection point in the field dependence of the magnetic moment whose shape resembles that of the present results. However, for the parameter sets they employ, the inflection point occurs at magnetic fields several times higher than the observed inflection point and it seems difficult to obtain a semi-quantitative agreement with experimental values of the size of the ordered moment, the transition temperature, and the inflection point, simultaneously.

We note in passing that a magnetic field applied along the c axis induces a localized ferromagnetic component, resulting in a ferrimagnetic structure with two different moments on the uranium-ion sites. The magnitude of the ferromagnetic moment at the metamagnetic transition can be estimated to $0.32 \mu_B$, based on an extrapolation of low-field polarized neutron diffraction measurements [22].

The magnetic excitations at the local minima $\mathbf{Q}=(1,0,0)$ and $(1.4,0,0)$ of the dispersion curve have been measured at low temperature ($T = 2$ K) for magnetic fields up to 17 T using the FLEX spectrometer of HMI. These data have been combined with earlier measurements at $H \leq 12$ T using the IN14 spectrometer of ILL [14]. The results are shown in Fig. 3. The measurements on IN14 have better statistics, due to higher flux and a larger sample volume (the sample volume is very limited in the 17 T magnet). Instrumental resolution effects in combination with the steep dispersion introduce an asymmetric high-energy tail of the sharp excitation at the magnetic zone center $\mathbf{Q}=(1,0,0)$. In such a case, the excitation energy can be extracted by fitting the convolution of a Gaussian function, which describes the energy resolution, with an exponential saw tooth function $\exp[-(E - \Delta)/\epsilon]$, which describes how the higher-energy excitations are coupled into the measured spectrum via the finite Q resolution. The field dependence of the parameter ϵ , which has no physical meaning, was determined from measurements at high energy transfers on IN14, and extrapolated to higher field values. This method gives a very accurate description of the measured spectra, as shown in Fig. 3(a), and the obtained excitation energy Δ lies, as expected, at approximately the half-height of the leading (low-energy) edge of the measured intensity. The peak shape of

the excitation at the second minima, $\mathbf{Q}=(1.4,0,0)$, is symmetric [see Fig. 3(b)] because of the smaller dispersion, and a Gaussian fit is sufficient to extract the corresponding excitation energy ∇ .

The magnetic field dependence of the gap energies Δ and ∇ are shown in Fig. 4. The gap at the AFM zone center increases rather strongly with applied field, without any sign of saturation at higher fields. It is well described by

$$\Delta(H) = [\Delta_0^2 + (bH)^2]^{1/2} \quad (5)$$

with $\Delta_0 = 1.59$ (2) meV and $b = 0.114$ (1) meV/T. The gap $\nabla = 4.51$ (2) meV at $\mathbf{Q}=(1.4,0,0)$ is nearly constant, with only a slight linear decrease in energy with increasing field, with slope 5.4 ± 2.7 μ eV/T. It is interesting to note that the high-field extrapolation of the energy gaps $\Delta(H)$ and $\nabla(H)$ crosses at a field close to the metamagnetic transition H_C . This is very interesting in the light of the appearance of a new high-field phase above 36.1 T [23], which could be due to Fermi surface effects requiring a different nesting vector, such as the one corresponding to $\mathbf{Q}=(1.4,0,0)$.

Specific heat [10] and electrical resistivity [24] data taken with a magnetic field $H \parallel c$ up to 17.5 and 16 T, respectively, have been analyzed in terms of a field-dependent energy gap related to the excitation gap observed in neutron scattering measurements. In both cases, the Néel temperature and the gap energy decrease quadratically with the field, following the relations $T_N(H) = T_N(0)[1 - (H/H_0)^2]$ and $\Delta(H) = \Delta(0)[1 - (H/H_0)^2]$, respectively, with H_0 being close to the metamagnetic field. Very recent specific-heat measurements at fields up to 45 T [23] show that T_N vanishes at a critical field of 35.9 T. The field dependence of T_N is essentially quadratic. Thermal expansion data [25] with a magnetic field $H \parallel c$ up to 25 T have been fitted by the empirical function $A \exp(-\Delta/T)$ for the magnetic contribution. A quadratic decrease of Δ and T_N was found with an extrapolated critical field of 37 T and a gap energy of 115 K at zero field.

The important point is that the energy gap extracted from different bulk measurements has the opposite magnetic field dependence to the gap Δ observed directly by neutron scattering at the AFM zone center. This low-energy gap actually *increases* strongly with field. The second gap ∇ shows a very slight linear decrease, much slower than a quadratic decrease observed in macroscopic measurements. While specific heat and electrical resistivity measure some average gap energy, it is difficult to imagine how an average of a strongly

increasing low-energy gap and a nearly field-independent high-energy gap would give rise to a decreasing gap. It is therefore clear that the gap from bulk measurements is unrelated to the gap(s) in the magnetic excitation spectra. This is in stark contrast to the transition temperature, which has the same field dependence in specific heat [10, 23, 25], electrical resistivity [23, 24], thermal expansion [25], and neutron scattering [4, 26] measurements. Unfortunately, due to the difficulty to measure T_N accurately for small moments with neutron scattering, there are no systematic neutron measurements of $T_N(H)$. It is interesting to note that the height of the anomalies in the specific heat ΔC_p [10] and resistivity $\Delta\rho$ [24] as well as the intensity of the magnetic gaps Δ and ∇ (see Fig. 3) are field independent.

In conclusion, our neutron scattering measurements on the heavy-fermion superconductor URu₂Si₂ show that the magnetic field dependence of the antiferromagnetically ordered moment m has a clear inflection point at ~ 7 T and remains finite even at 17 T for fields applied along the c axis. This strongly suggests the existence of a hidden order parameter ψ that breaks time-reversal symmetry. Symmetry arguments show that the hidden order parameter belongs to the same irreducible representation A_{2g} as m and corresponds to (ordinary) dipolar moments of the *conduction electrons* or triple-spin correlators of the $5f$ electrons. Our neutron scattering measurements of the (gapped) magnetic excitations show that they are *not* related to the gap extracted from e.g. specific heat measurements, since the magnetic field dependence of these gaps is completely different. Instead, it is the hidden order parameter that gives rise to the large anomaly in specific heat and other macroscopic properties. The extrapolated crossing of the two magnetic gaps might explain the new phase recently discovered [23] above the metamagnetic transition.

We acknowledge fruitful discussions with G. Amoretti, N.H. van Dijk, J. Flouquet, I. Fomin, V.P. Mineev, K. Miyake, P. Santini, J. Schweizer, M.B. Walker, and M.E. Zhitomirsky. A.A. Menovsky, P. Lejay, and A.D. Huxley contributed to sample growth and preparation. We thank M. Jaime for giving us access to his high-field specific-heat data prior to publication.

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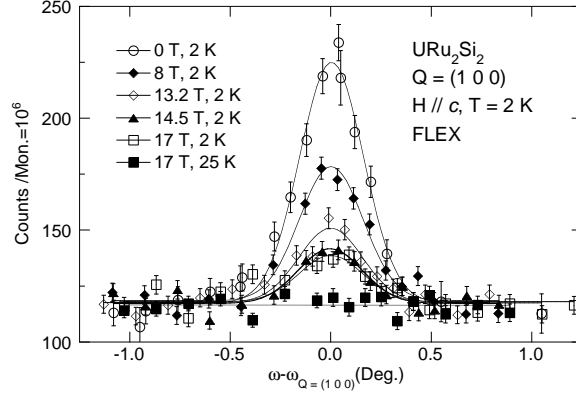


FIG. 1: Rocking scans of the AFM Bragg peak for different values of the magnetic field. The lines are Gaussian fits.

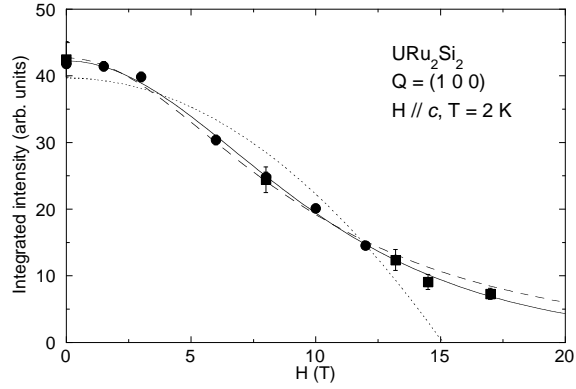


FIG. 2: Integrated intensity of the AFM Bragg peak as a function of magnetic field. Squares (circles) are from FLEX (IN14). The dotted, solid, and dashed lines correspond to fits given by Eqs. (1), (3), and (4), respectively.

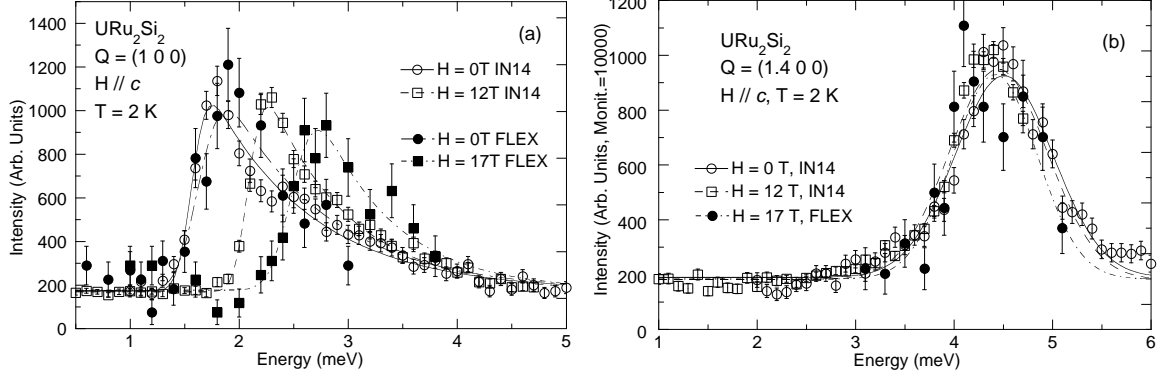


FIG. 3: Energy scans of the magnetic excitations at the two local minima $Q=(1,0,0)$ (a) and $Q=(1.4,0,0)$ (b) for magnetic fields of 0, 12, and 17 T. The intensity scales for the data on the IN14 and FLEX spectrometers are scaled to coincide. The lines are fits described in the text.

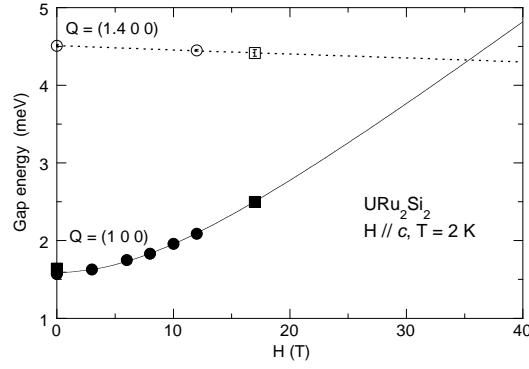


FIG. 4: The gap energies at $Q=(1,0,0)$ (solid symbols) and $Q=(1.4,0,0)$ (open symbols) as a function of magnetic field measured on IN14 (circles) and FLEX (squares). The solid line is a fit to Eq. (5) and the dotted line is a linear fit.